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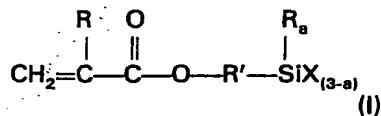
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(54) A Method of Bonding a Silicone Rubber to a Substrate Surface

(57) An acryloxyalkylsilane is mixed into a "high viscosity" i.e. an organic peroxide-vulcanized silicone rubber. The rubber is a polydiorganosiloxane containing organic radicals selected from methyl, vinyl, phenyl, and 3,3,3-trifluoropropyl.

The acryloxyalkylsilane has the formula (I):



in which:

R is selected from hydrogen and methyl radical,
 R' is an alkylene radical of from 1 to 4 carbon atoms inclusive,
 X is a radical selected from alkoxy radicals of from 1 to 3 carbon atoms inclusive, and the acetoxy radical, and a is from 0 to 2 inclusive.

The mix, which also includes the organic peroxide vulcanizing agent, silica fillers and an anti-crepe hardening agent, is applied to a surface of a substrate and heated to vulcanization temperature to bond the silicone rubber composition to the substrate.

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SPECIFICATION

A Method of Bonding a Silicone Rubber to a Substrate Surface

This invention relates to a method of bonding a silicone rubber to a substrate surface. In particular, the invention is concerned with a method of bonding a heat-activated organic peroxide-vulcanized silicone rubber to the surface of a substrate during the vulcanizing of the rubber.

Certain uses for silicone rubber such as shock mounts and metal-enclosed shaft seals required that the rubber be firmly bonded to the surface of the substrate.

Two general methods are used for bonding silicone rubber to the surfaces of substrates. The silicone rubber can be formed to shape and vulcanized, as in a mould, then bonded to a substrate surface with an adhesive. Alternatively, the unvulcanized silicone rubber stock can be applied to the substrate surface and then vulcanized.

In either case, most types of substrate surfaces must be carefully cleaned and then treated with special primers in order to obtain satisfactory adhesion of the vulcanized silicone rubber to the substrate surface. The priming of the substrate surface before the bonding step is a costly and time-consuming operation that is not necessary in the method of this invention.

A majority of the commercial primers presently available are activated when applied to a substrate surface by the moisture in the air. The variability of the drying and hydrolysing conditions due to day-to-day differences in the humidity in the air can lead to variability of results.

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A different method of obtaining adhesion to a substrate surface is through the addition of adhesion additives to the unvulcanized silicone rubber stock. U.S. Patent No. 4,033,924 to Mine *et al.* discloses a heat-curable organopolysiloxane composition containing an organosilicon compound having at least one $A(R'O)_2Si$ group and at least one alkyl, low molecular weight alkenyl group, or hydrogen atom bound to silicon, A is a monovalent epoxy-containing hydrocarbon group and R' is a low-molecular weight alkyl group.

25 An improved method of obtaining bonding of a heat-activated, organic peroxide vulcanized silicone rubber to substrate surfaces has been developed. A silicone rubber composition of the type commonly known as "high viscosity", that is, vulcanized through the use of organic peroxides, is used as the basic material. The first step of the method is mixing into the silicone rubber composition an acryloxyalkylsilane. This modified composition is then formed into the desired shape in contact with the

30 cleaned substrate surface to which it is to be bonded. The modified composition is then heated to vulcanize it while it is in contact with the substrate surface. Bonding of the modified composition to the substrate surface takes place during the vulcanization step.

This invention relates to a method for improving the bonding of a vulcanized silicone rubber to a substrate surface comprising:

35 (a) mixing 100 parts by weight of silicone rubber base consisting essentially of a polydiorganosiloxane containing organic radicals selected from vinyl, methyl, phenyl and 3,3,3,-trifluoropropyl and comprising reinforcing silica filler and an anticrepe-hardening agent; from 0 to 150 parts by weight of siliceous extending filler with an average particle size of less than 25 micrometres and a surface area of less than 50 m²/g; from 0.1 to 5 parts by weight of organic peroxide vulcanizing agent suitable for vulcanizing the silicone rubber base; and from more than 0.1 to 1.5 parts by weight of an acryloxyalkylsilane of the formula (7):



45 "in which R is selected from hydrogen and methyl radicals, R' is an alkylene radical of from 1 to 4 carbon atoms inclusive, X is a radical selected from alkoxy radicals of from 1 to 3 carbon atoms inclusive, and the acetoxy radical, and a is from 0 to 2 inclusive", to yield a curable silicone rubber composition;

(b) forming a composition wherein the curable silicon rubber composition contacts a surface of a substrate, and, thereafter,

(c) heating the combination to a temperature sufficiently high to vulcanize the composition, thereby yielding a vulcanized silicone rubber bonded to the substrate surface.

50 The silicone rubber base used in the present invention can be any mixture of polydiorganosiloxane and reinforcing silica filler including types commercially available. The polydiorganosiloxane of this invention contains organic radicals selected from methyl, vinyl, phenyl, and 3,3,3-trifluoropropyl, said radicals being bonded to the silicon atoms of the polydiorganosiloxane. The polydiorganosiloxanes are commonly of a viscosity of from 1000 Pa's up to and including non-flowing gums. These polydiorganosiloxanes are well known in the art and are commercially available.

55 A silicone rubber base contains a reinforcing silica filler to improve the physical strength of the polymer. Reinforcing silica fillers have surface areas of from 150 to above 400 m²/g. These reinforcing silica fillers are well known in the art and can be obtained commercially. The reinforcing filler can be untreated, treated, or treated *in situ* during the manufacture of the silicone rubber base. The treated 60 reinforcing silica fillers can be treated by any of the conventional methods described in the prior art,

wherein the treating agents include organosilanes, organosiloxanes and silazanes. The amount of reinforcing filler can vary from 10 to as much as 100 parts by weight, the usual amount varying from 15 to 75 parts by weight per 100 parts by weight of the polydiorganosiloxane.

A silicone rubber base may also contain anti-crepe hardening agents. These anti-crepe hardening agents are used to reduce the reaction between the polydiorganosiloxane and the reinforcing silica that causes the base to become harder or pseudo-vulcanized. Such a reaction can cause the base to become to "nervy" to be of further use. 5

Suitable anti-crepe hardening agents are well known in the art. They can be such additives as hydroxyl-endblocked, short-chain polydimethylsiloxane fluids. If the reinforcing filler is treated as 10 discussed above, the silicone rubber base may not need an additional anti-crepe hardening agent. 10

The silicone rubber base may also contain minor amounts of additives to improve, among other things, the heat stability, handling, compression set and oil resistance. A single silicone rubber base may be used to obtain the desired range of physical properties for the cured silicone rubber.

In use, a silicone rubber base may be extended with an extending filler to increase the bulk of the 15 composition. This helps to lower the cost of the finished part as the extending fillers are much lower in cost than the silicone rubber base. When a silicone rubber base is extended with an extended filler such as ground quartz, the tensile strength of the cured composition is lower than that of the starting base. The amount of tensile strength lost is dependent upon the relative amounts of base and extending filler used as well as the exact nature of both ingredients. 15

20 The addition of an extending filler may also lower the bond strength of a composition intended to be bonded to a substrate surface. The method of this invention provides a means whereby compositions containing large amounts of extending filler can still be successfully bonded to substrate surfaces. As higher levels of extending filler are added, it becomes more difficult to achieve a satisfactory bond to a substrate surface. The maximum amount of extending filler that can be used and 25 still give satisfactory bonding to a substrate surface will depend upon the nature of the silicone rubber base and the extending filler used. The maximum is substantially 150 parts by weight of extending filler per 100 parts by weight of silicone rubber base. 25

The siliceous extending fillers used with silicone rubber bases are finely ground particles of heat-stable inorganic materials with an average particle size of under 25 micrometres. The finest extending 30 fillers approach a particle size and configuration such that they have a surface area as high as 50 m²/g. Examples of siliceous extending fillers include ground quartz, diatomaceous earth and glass. 30

About 25 parts by weight of extending filler per 100 parts by weight of silicone rubber base are necessary significantly to lower the cost of the composition. The preferred siliceous extending fillers for use with the present invention are ground quartz and diatomaceous earth with the most preferred 35 filler being ground quartz with an average particle size of substantially 5 micrometres. 35

The composition of this invention contains an organic peroxide vulcanizing agent suitable for vulcanizing the polydiorganosiloxane in the silicone rubber base. When the polydiorganosiloxane does not contain any vinyl radicals, it must be vulcanized with organic peroxides that are efficient in causing 40 reactions in such polydiorganosiloxanes. Such organic peroxides are labelled "non-vinyl specific" and are represented by such well-known organic peroxides as benzoylperoxide, dicumylperoxide and 2,4-dichlorobenzoylperoxide. When the polydiorganosiloxane contains vinyl radicals, it can be vulcanized with either "non-vinyl specific" or "vinyl specific" organic peroxides. Diteriary-butyl peroxide and 2,5-bis-(tert-butylperoxy)-2,5-dimethyl-hexane are representative of the vinyl specific organic peroxides. All these organic peroxide vulcanizing agents and their properties are well known in the art. The 45 properties of the vulcanized silicone rubber can be altered by the type and amount of vulcanizing agent used to vulcanize the composition. Typical changes due to such choices are well recognized in the art. The vulcanizing agent can be present in amounts of from 0.1 to 5 parts by weight per 100 parts by weight of silicone rubber base, preferably from 0.5 to 2.0 parts by weight. 45

The critical component of the composition used in the method of this invention is an 50 acryloxyalkylsilane of the formula (I): 50



in which R is selected from hydrogen and methyl radicals, R' is an alkylene radical of from 1 to 4 inclusive carbon atoms, X is a radical selected from alkoxy radicals of from 1 to 3 carbon atoms inclusive, and the acetoxy radical, and a is from 0 to 2 inclusive. The silane is preferred where R is a 55 methyl radical, a is 0, and X is a methoxy radical or acetoxy radical. The most preferred silane is gamma-methacryloxypropyltrimethoxysilane because of its efficiency in causing the vulcanized silicone rubber to bond to a substrate surface against which the silicone rubber has been vulcanized. 55

The acryloxyalkylsilanes used in this invention are known in the art. They are disclosed in U.S. Patent No. 3,567,497 by Pluedemann and Clark which describes the silanes and their method of manufacture. The preferred gamma-methacryloxypropyltrimethoxysilane is commercially available. 60

The compositions of this invention bond to a substrate surface when the composition is cured

while in contact with the substrate surface during the vulcanization of the composition. In order to obtain bonding, it is necessary to use at least 0.1 part by weight of silane per 100 parts by weight of silicone rubber base. The exact amount of silane necessary to obtain bonding and the optimum property profile of the cured silicone rubber composition can be easily determined by simple

5 experimentation. The results will depend upon the silicone rubber base selected, the kind and amount of extending filler used, the kind and amount of vulcanizing agent used, and the nature of the substrate surface to be adhered to. The preferred amount of silane is from above 0.1 to 1.0 part by weight per 100 parts by weight of silicone rubber base. The addition of more than 1.5 parts by weight of silane will not improve the adhesion and will begin adversely to affect the physical properties of the cured 10 silicone rubber composition.

10 The mixing step of this invention used to prepare the composition can be any suitable means that will lead to a homogeneous mixture of the several components. Methods of mixing that are common in the silicone rubber art and which are suitable for this invention include mixing with a dough mixer, a rubber compounding mill or a Banbury mixer. The order of mixing is not critical. Ordinarily, the silicone 15 rubber base is placed in the mixer the extending filler and silane are added and mixed until homogeneous, the vulcanizing agent is added and mixing continued until the mixture is homogeneous. Any additional additives such as heat stability additives, antioxidants, processing aids, pigments, and so on are ordinarily added before the vulcanizing agent.

15 The compositions can be formed to the desired configuration by any of the well-known methods 20 of forming elastomeric curable compositions such as press moulding, injection moulding, calendering and extruding, both supported and unsupported. Since the compositions bond without primers, special precautions must be taken during the vulcanizing operations to ensure that the vulcanized composition adheres only to surfaces where adhesion is desirable. The surfaces of press plates or moulds, for instance, must well be coated with a suitable release agent.

25 Suitable release agents for the method of this invention are heavy coats of a 2 to 5 percent by weight solution of detergent in water, or, more preferably, a coating of fluorocarbon mould release agent. For flat surfaces, a sheet of polytetrafluoroethylene is satisfactory.

30 The formed compositions of this invention can be vulcanized by any suitable means that will cause decomposition of the organic peroxide vulcanizing agent. Heating is the preferred method. The 30 time and temperature necessary to cause vulcanization of the composition is dependent upon the organic peroxide vulcanizing agent chosen, the method of heating, the method of shaping the composition to the desired configuration, and the thickness of the part. The temperature that is appropriate for a given set of conditions is well known in the silicone rubber art. Typical temperatures are from 110°C. to 175°C. for moulding operations, to as high as 300°C. for the ovens used in 35 continuous hot air vulcanization operations.

35 The method of this invention is useful for making silicone rubber articles that are bonded to a substrate surface. Examples of such articles are metal enclosed shaft seals, shock mounts, rolls, and various types of fabric-reinforced articles such as tubing, tapes and diaphragms.

40 The following examples are included for illustrative purposes only and should not be construed as 40 limiting the invention which is properly delineated by the appended claims. All parts are parts by weight.

Example 1

45 A series of samples were made to evaluate the bonding characteristics of stock containing an acryloxyalkylsilane as compared with the same stock without the silane.

45 A stock was compounded consisting of:

(a) 100 parts of commercial silicone rubber base described as a vinyl-containing silicone rubber designed for compounding general purpose silicone rubber stock. The base was translucent with a specific gravity after curing of 1.09. The base consisted of a vinyl-containing polydimethyl-siloxane, a reinforcing fume silica, and a hydroxyl-endblocked, polydimethylsiloxane fluid to prevent crepe-

50 hardening of the base;

(b) 50 parts of ground quartz with an average particle size of 5 micrometres,

(c) 2 parts of iron oxide paste; and

(d) 1 part of organic peroxide vulcanizing agent consisting of 50 weight percent 2,5-bis-(tert-butylperoxy)-2,5-dimethylhexane dispersed on an inert carrier powder.

55 0.5 Part of gamma-methacryloxypropyltrimethoxysilane based on 100 parts of the silicone rubber base was added to a portion of this stock.

55 Each stock was then calendered onto a piece of glass fiber fabric to a total thickness of 0.5 mm. The silicone rubber surface of the calendered fabric was then press-moulded against the cleaned surface of metal panels as shown in Table I. The surface of the metal panels was cleaned by wiping thoroughly with chlorothene and then with acetone. Two pieces of the calendered fabric were also moulded against each other with the rubber surfaces in contact. The mouldings were for 10 minutes at 171°C.

60 After moulding, each sample was cut into 25.4 mm wide strips. The calendered fabric strips were then pulled from the various substrate surfaces using a standard tensile testing machine with a rate of

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50.8 mm per minute. The strips were pulled from the substrate surface at an angle of 180°. The glass fiber fabric samples were pulled from one another at a total angle of 180° or at 90° each at the point of peeling apart.

The method of failure was noted for each sample. If there was no adhesion, it was recorded as 5 zero percent cohesive failure. If the sample failed by tensile failure within the rubber itself, it was recorded as 100 percent cohesive failure. 5

The results are shown in Table I below. The addition of the silane to the silicone rubber stock greatly improved the adhesion to all of the substrate surfaces tested.

Table I

10	Amount of Silane	Substrate	Adhesion kN/m	Failure Type % cohesion	10
	none	Aluminum	0.05		
15	none	C.R. Steel	0.13	0	
	none	Stainless Steel	0.13	0	15
	none	Glass-Fibre Fabric	0.56	5	
20	0.5 part	Aluminium	2.45	100	
	0.5 part	C.R. Steel	2.62	100	20
	0.5 part	Stainless Steel	1.75	50	
	0.5 part	Glass-Fibre Fabric	2.80	80	

25 **Example 2**

A different commercial silicone rubber base was used to prepare samples in the same manner as in Example 1.

A stock was compounded consisting of:

(a) 100 parts of a commercial silicone rubber base similar to that of Example 1 but with a higher 30 loading of reinforcing silica (the specific gravity of the translucent base was 1.10);
 (b) 2 parts of iron oxide paste; and
 (c) 1 part of the organic peroxide vulcanizing agent of Example 1.

0.5 Part of the silane used in Example 1 based on 100 parts of stock was added to a portion of the stock.

35 Test samples were prepared and tested in the same manner as described in Example 1. The results are recorded in Table II below. The addition of the silane to a silicone rubber stock not containing extending filler greatly improved the adhesion to all the substrate surfaces tested.

A sample of each type of substrate surface was primed with a commercial primer. The primer was used with an organic peroxide-catalysed, heat-cured, silicone rubber stock to bond without oven post-curing. A sample was prepared using the above stock of this Example without the silane added. The 40 samples were moulded and tested as described in Example 1. The results are recorded in Table II. The stock bonded with the primer did not give as high a bond strength as that of the stock bonded by adding the silane to the stock. The adhesion shown by the glass-fibre fabric samples is partially due to mechanical trappings of the rubber into the rough surface of the fabric. The amount of cohesive failure 45 is also more difficult to judge due to the very uneven nature of the surface.

Table II

50	Amount of Silane	Substrate	Adhesion kN/m	Failure Type % cohesion	50
	none	Aluminium	0.12		
		C.R. Steel	0.35	0	
		Stainless Steel	0.53	0	
		Glass-Fibre Fabric	0.88	0	
55	0.5	Aluminium	3.7	100	
		C.R. Steel	3.3	100	55
		Stainless Steel	3.2	95	
60		Glass-Fibre Fabric	3.8	80	60

		Table II (cont.)			
	Amount of Silane	Substrate	Adhesion kN/M	Failure Type	
5	1.0	Aluminium	4.3	100	5
		C.R. Steel	4.0	100	
		Stainless Steel	3.4	100	
		Glass-Fibre Fabric	4.2	20	10
		Aluminium	0.88	40	
		C.R. Steel	1.1	20	
10	primer	Stainless Steel	0.96	50	
		Glass-Fibre Fabric	4.1	15	15

Example 3

Different types of additives were mixed with a commercial silicone rubber base to compare their usefulness in improving bonding to glass-fibre fabric.

A stock was compounded consisting essentially of:

20 (a) 100 parts of a commercial silicone rubber base designed to give a 70 durometer, high tensile strength product. The specific gravity of the base was 1.21;

(b) 50 parts of ground quartz with an average particle size of 5 micrometres; and

(c) 1 part of the organic peroxide vulcanizing agent of Example 1.

25 Portions of the stock were mixed with 1 part of the additives, detailed below, on a 2-roll mill.

Each sample was then moulded under low pressure in a press against a coarse weave, untreated glass-fibre fabric for 10 minutes at 171°C. The samples were then evaluated by pulling the cured silicone rubber stock and the glass-fibre fabric apart. The results are recorded in Table III, below.

Sample 1 had no additive.

30 Sample 2 was a mixture of a trimethylsiloxy-endblocked polymethylhydrogensiloxane with a silicon-bonded hydrogen atom content of about 1.6 weight percent and ethylpolysilicate. This mixture is known to give bonding.

Sample 3 was gamma-glycidoxypropyltrimethoxysilane. This epoxy-functional silane is used to aid bonding with many different types of polymer.

35 Sample 4 was gamma-methacryloxypropyltrimethoxysilane. The results show that the silane of this invention gave superior bonds as compared with the other additives tested.

Table III

	Sample	Result	
40	1 Blank	weak mechanical bond, 0% cohesive failure	40
	2 Comparative Example	some bonding, 0% cohesive failure	
	3 Comparative Example	stronger than 2, 0% cohesive failure	
45	4 This invention	strong bond, 100% cohesive failure	45

Example 4

A series of samples were made to evaluate the effect of adding acryloxyalkysilane on the physical properties of the cured silicone rubber composition.

50 A stock was compounded consisting of 100 parts of the commercial silicone rubber base of Example 1, 100 parts of the ground quartz extending filler of Example 1, and 1 part of the organic peroxide vulcanizing agent of Example 1.

Portions of the above stock were then mixed with gamma-methacryloxypropyltrimethoxysilane in the amounts shown in Table IV below for 100 parts of base.

55 Each portion was moulded into test slabs in a press, using aluminium plates treated with a commercial soap-type release agent specified for use with heat-cured silicone rubber. The moulding was for 10 minutes at 171°C. The samples containing 0.5 and 1.0 part of the silane were very difficult to remove from the aluminium plates, even though the plates were coated with a release agent.

60 The physical properties of the slab were determined in accordance with the procedures described by ASTM-412 for tensile strength and elongation, by ASTM-D625, die B tear strength, and by ASTM-D2240 for durometer, type A. The measured physical properties were shown in Table IV in which the tensile strength is recorded in megapascals (MPa) and the tear strength is recorded in kilonewtons per meter (kN/M).

The addition of the silane to the compound stock containing a siliceous extending filler did not harm the physical properties. It caused a significant improvement in the tensile strength.

Table IV

	Amount of Silane parts/100 parts base	Durometer	Tensile Strength MPa	Elongation %	
5	none	60	3.1	400	5
	0.1	62	6.8	200	
	0.5	62	6.9	180	
10	1.0	63	6.0	150	10

Example 5

A series of samples were made to evaluate the level of acryloxyalkylsilane needed in a vulcanized silicone rubber stock to bond to glass-fibre fabric.

A stock was compounded consisting of 100 parts of the commercial silicone rubber base of 15 Example 3, 25 parts of the ground quartz of Example 3, 1 part of a commercial heat-stability additive, and 1 part of the organic peroxide vulcanizing agent of Example 1. 15

Portions of the stock were then mixed with the amounts of gamma-methacryloxypropyltrimethoxysilane shown in Table V below based on 100 parts of the silicone rubber base.

20 Each portion was then calendered onto style 1528 glass fibre fabric to a total thickness of 0.5 mm. Test samples were prepared by placing pieces of each calendered sample together such that the sample was 4 plies thick. The two centre plies were placed rubber face to rubber face. Each sample was moulded for 10 minutes at 171°C. in a press under light pressure to vulcanize the stock and to bond the pieces together. 20

25 The samples were then evaluated by pulling the pairs of plies apart in a standard test machine at a rate of 50.8 mm per minute causing the plies to separate at the centre interface where two rubber layers were together. The plies were pulled from one another at a total angle of 180°C or at 90° each at the point of peeling apart. 25

The method of failure was noted for each sample. The results are shown in Table V.

30 The addition of the silane to the stock used in this method of bonding improved the bond over that obtained with no silane. The failure at the 0.1 part level appeared to be an adhesive failure, but the higher peel strength shows that some adhesion must have been taking place. The lower peel strengths for the samples with 0.75 part silane and 1.0 part silane are probably due to the higher modulus of those stocks and its effect on the geometry of the failure point as the pieces are pulled apart. 30

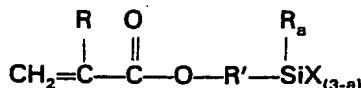
Table V

	Amount of Silane	Adhesion kN/m	Failure Type	
35	None	0.35	0%	35
			cohesion	
40	0.1	0.78	10	40
	0.25	1.3	100	
	0.5	1.1	100	
	0.75	0.91	100	
	1.0	0.96	100	

45 Claims

1. A method of improving the bonding of a vulcanized silicone rubber to a substrate surface comprising:

(a) mixing 100 parts by weight of silicone rubber base consisting essentially of a polydiorganosiloxane containing organic radicals selected from methyl, vinyl, phenyl and 3,3,3-trifluoropropyl and comprising reinforcing silica filler and an anticrepe-hardening agent; with from 0 to 50 parts by weight of siliceous extending filler with an average particle size of less than 25 micrometres and a surface area of less than 50 m²/g; from 0.1 to 5 parts by weight of organic peroxide vulcanizing agent suitable for vulcanizing the silicone rubber base; and from above 0.1 to 1.5 parts by weight of an acryloxyalkylsilane of the formula:



"in which R is selected from hydrogen and methyl radical, R' is an alkylene radical of from 1 to 4

inclusive carbon atoms, X is a radical selected from alkoxy radicals of from 1 to 3 carbon atoms inclusive, and the acetoxy radical, and a is from 0 to 2 inclusive" to yield a curable silicone rubber composition:

(b) forming a combination wherein the curable silicone rubber composition contacts a surface of

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5 a substrate; and, thereafter,

(c) heating the combination to a temperature sufficiently high to vulcanize the composition, thereby producing a vulcanized silicone rubber bonded to the substrate surface.

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2. The method of claim 1 wherein the siliceous extending filler is selected from ground quartz and diatomaceous earth, the organic peroxide vulcanizing agent is present in an amount of from 0.5 to 2.0 parts by weight, and the acryloxyalkylsilane is gamma-methacryloxypropyltrimethoxysilane.

10

3. The method of claim 1 or claim 2 wherein the substrate surface is metal or glass.

4. A method according to claim 1, substantially as herein described with reference to any of the specific examples.

15

5. A vulcanized silicone rubber bonded to a substrate surface by the method of any
15 of claims 1 to 4.

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